

2003年11月14日 16時01分

栗原国際特許事務所

NO. 6358 P. 2



PATENT APPLICATION  
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN THE APPLICATION OF  
KAZUMA NIUME, ET AL.

SERIAL NO.: 09/889,395

EXAMINER: CALLIE E. SHOSH

FILED: JULY 17, 2001

ART UNIT: 1714

TITLE: COATING SOLUTION FOR FORMING TRANSPARENT CONDUCTIVE TIN OXIDE  
FILM, METHOD FOR PRODUCING TRANSPARENT CONDUCTIVE TIN OXIDE FILM, AND  
TRANSPARENT CONDUCTIVE TIN OXIDE FILM

RECEIVED  
NOV 25 2003  
TC 1700

DECLARATION UNDER 37 C.F.R. §1.132

I, Takashi Uchida, hereby declare THAT:

1. I received my bachelor's, master's and doctorate degrees in Engineering from Tokyo Institute of Technology. The degrees were awarded in March of 1974, 1976 and 1979, respectively. During that time I was a member of the faculty of Engineering and Department of Chemical Engineering;

2. In April of 1979, I was appointed a Research Associate in the department of Chemical Engineering at Tokyo Institute of Technology. In August of 1990, I was appointed as Associate Professor in the Department of Chemical Engineering;

3. From May of 1999 to present, I have been employed as a Chief Chemist of Toyo Gosei Co., LTD.

I am an author or co-author of many technical publications, including:

A) Chemical Diffusion Coefficient of Lithium in Carbon Fiber, T. Uchida, Y. Morikawa, H. Ikuta, M. Wakiyara, and K. Suzuki, J. Electrochem. Soc., 143, 2606 (1996).

B) Amorphous  $\text{MoS}_2$  as the Cathode of Lithium Secondary Batteries, Y. Miki, D. Nakazato, H. Ikuta, T. Uchida, and M. Wakiyara, J. Power Sources, 54,

2003年11月14日 16時01分

栗原国際特許事務所

NO. 6358 P. 3

508 (1995).

C) Structure, Structural Defects, and Reactions in a Nickel Chevrel Phase Sulfides: A High-Resolution Electron Microscope Study, Z. C. Kang, L. Eyling, H. Minode, T. Uchida, and M. Wakihara, J. Solid State Chem., 111, 58 (1994) .

D) High Ionic Conductivity in Lithium Lanthanum Titanate, Y. Inaguma, Chen. Liqun, M. Itoh, T. Nakamura, T. Uchida, H. Ikuta, and M. Wakihara, Solid State Commun., 86, 689 (1993).

E) A Rechargeable Lithium Battery Employing Cobalt Chevrel Phase Compound as the Cathode, S. Yamaguchi, T. Uchida, and M. Wakihara, J. Electrochem. Soc., 138, 687 (1991).

F) Nickel-Molybdenum Sulfide  $\text{Ni}_2\text{Mo}_6\text{S}_8$  as the Cathode of Lithium Secondary Batteries, T. Uchida, Y. Tanjo, M. Wakihara, and M. Taniguchi, J. Electrochem. Soc., 137, 7 (1990).

G) Electrical Conductivity of the  $\text{CaF}_2\text{-NaF}$  System, M. Natsuhara, T. Uchida, and M. Wakihara, Solid State Ionics, 40/41, 874 (1990).

H) A Rechargeable Lithium Battery Employing Iron Chevrel Phase Compounds ( $\text{Fe}_{1.25}\text{Mo}_6\text{S}_8$ ) as the Cathode, M. Wakihara, T. Uchida, K. Suzuki, and M. Taniguchi, Electrochim. Acta., 34, 867 (1989).

I) Ionic Conductivity of  $\text{Li}_{4-2x}\text{Mg}_x\text{PO}_4$ , M. Wakihara, T. Uchida, and T. Gohara, Solid State Ionics, 31, 17 (1988).

I am an inventor of the invention disclosed in the above-identified patent application.

I had performed certain experiments to show the differences between the above application, Serial No. 09/889,395 and the cited reference, USP

2003年11月14日 16時02分

栗原国際特許事務所

NO. 6358 P. 4

6,066,442 to Kurachi et al. I reported the experiments and the results below:

(Experiment 1) Performed according to the teachings of Kurachi et al.

65 g of stannic chloride hydrate was uniformly dissolved in 2000 cc an aqueous water. Next the resulting solution was boiled to thereby yield the precipitate. The precipitate was centrifuged twice for washing. The resulting precipitate was added to 2000 cc of distilled water and dispersed. In addition, 40cc of 30% aqueous ammonia was added to the mixture. After 5 minutes, the resulting mixture was heated in a water bath for 30 minutes and particles grew. This solution in which the particles are dispersed was allowed to stand for 24 hour at ambient temperature. However, the particles are dispersed but not dissolved in the solution. The pH of the end solution was 9.50.

(Experiment 2) Performed according to the present invention

65 g of stannic chloride hydrate was uniformly dissolved in 2000 cc an aqueous water. Next 30 % aqueous ammonia was added to the solution until the pH value increased up to 7.0, to thereby yield the precipitate. The precipitate was centrifuged twice for washing. The resulting precipitate was added to 2000 cc of distilled water and dispersed. In addition, 40cc of 30% aqueous ammonia was added to the mixture. The resulting mixture was allowed to stand for 2 hours at ambient temperature. The resulting solution was clear and contained no particles. The pH of the end solution was 10.45.

(Test Experiment 1)

Poly dimethylacrylamide (PDMAA) was mixed with the resulting solution of Experiment 1 so as to contain 0.3 wt% of PDMAA. A silica-coated soda-lime glass substrate was coated with the resulting solution by means of spin coating, then the substrate was calcinated at 550°C for 1 hour to form transparent SnO<sub>2</sub> film. Grains of the size 0.5 through 1 μm in diameter were observed on the surface by atomic force microscope (AFM) (Seiko Instruments Inc.; Nanopics1000) (See Fig. 1A). The

2003年11月14日 16時02分

栗原国際特許事務所

NO. 6358 P. 5

amplitude of asperities on the surface in the observed film was found to be around 30 nm (See Fig. 1B: cross-sectional view taken along blue line of Fig. 1A ).

(Test Experiment 2)

Transparent  $\text{SnO}_2$  film was formed in the same manner as shown in Test Experiment 1 using the resulting solution of Experiment 2. No distinct grain was observed on the surface by AFM (See Fig. 2A). The amplitude of asperities on the surface in the observed film was found to be around 10 nm (See Fig. 2B: cross-sectional view taken along blue line of Fig. 2A).

(Conclusion)

The above results show that the solution according to Kurachi et al. (Experiment 1) is a dispersed solution, while the solution of the present invention (Experiment 2) is a complete solution.

In addition, it is confirmed that the resulting film of the present invention (Test Experiment 2) is more flat than that of Kurachi et al. (Test Experiment 1).

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the present application or any patent issued thereon.

2003年11月14日 16時02分

栗原国際特許事務所

NO. 6358 P. 6

*Takashi Uchida*

Takashi Uchida

Date: November 14, 2003